Tuesday, May 12
Facility-specific Workshops

CNM Workshop 1
Nanophotonic Structures, Surfaces, and Composites for Radiative Control

Location: Bldg. 401, Room A5000

Organizers: Steve Smith (South Dakota School of Mines & Technology), Il Woong Jung (Nanofabrication & Devices Group, CNM), Zhe Fei (Nanophotonics Group, CNM))

The scope of this workshop encompasses all aspects of the photonic control of quantum systems, including the fabrication, characterization, simulation, and theory of composite photonic-electronic systems, that is, those specific materials and nano-structured composite systems which intentionally modify and/or exhibit control of light matter interactions as a consequence of their nanostructure. Examples are quantum emitters (e.g., molecules, ions or quantum dots) whose radiative properties are modified by their interactions with nano-optical structures, including photonic crystals, cavities, plasmonic surfaces and/or high-aspect ratio optical nano-structures. Development of improved optical materials, nanostructures, and composites and their applications will be emphasized as well as photonic crystals, micro-cavities, photonic control, plasmonics, fabrication, and the properties of photonic nano-structures.

8:30 – 8:45 Welcome & Introductory Remarks

8:45 – 9:25 Marko Loncar (Harvard University)
Quantum and Nonlinear Nanophotonics with Diamond

9:25 – 10:05 George Schatz (Northwestern University)
Plasmonic Arrays

10:05 – 10:35 Break

10:35 – 11:15 Yugang Sun (Argonne National Laboratory)
Interfacial Influence on Surface Plasmon Resonances in "Quantum-sized" Metal Nanoparticles

11:15 – 12:00 Zhiqun Lin (Georgia Tech)
Crafting Nanophotonic Structures

12:00 – 1:30 Lunch

1:30 – 2:10 Peng Zhang (University of Cincinnati)
SERS Inside Metal Nanoshell and Its Applications

2:10 – 2:50 Shaung Fang Lim (North Carolina State University)
Multifunctional Diagnostic, Nanothermometer and Photothermal Nano-devices

2:50 – 3:20 Break

3:20 – 4:00 Stephen Gray (Argonne National Laboratory)
Theoretical Studies of Hybrid Plasmon/Quantum-dot Systems

4:00 – 4:40	Nathaniel Stern (Northwestern University) *Photons in Flatland: Manipulating Light and Matter in Two-dimensional Nanomaterials*
4:40	Wrap-up and concluding remarks

WK1

Quantum and Nonlinear Nanophotonics with Diamond

Marko Loncar

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Diamond possesses remarkable physical and chemical properties, and in many ways is the ultimate engineering material — "the engineer's best friend!" For example, it has high mechanical hardness and large Young's modulus, and is one of the best thermal conductors. Optically, diamond is transparent from the ultra-violet to infra-red, has a high refractive index (n = 2.4), strong optical nonlinearity and a wide variety of light-emitting defects. Finally, it is biocompatible and chemically inert, suitable for operation in harsh environment. These properties make diamond a highly desirable material for many applications, including high-frequency micro- and nano-electromechanical systems, nonlinear optics, magnetic and electric field sensing, biomedicine, and oil discovery. One particularly exciting application of diamond is in the field of quantum information science and technology, which promises realization of powerful quantum computers capable of tackling problems that cannot be solved using classical approaches, as well as realization of secure communication channels. At the heart of these applications are diamond's luminescent defects — color centers — and the nitrogen-vacancy (NV) and silicon-vacancy (SiV) color center in particular. These atomic systems in the solid-state possesses all the essential elements for quantum technology, including storage, logic, and communication of quantum information.

I will review recent advances in nanotechnology that have enabled fabrication of nanoscale optical devices and chip-scale systems in diamond that can generate, manipulate, and store optical signals at the single-photon level. Examples include a room temperature source of single photons based on diamond nanowires [1] and plasmonic appertures [2], as well as single-photon generation and routing inside ring [3] and photonic crystal resonators [4]. Novel, fabrication technique [5] — *angled-etching* — suitable for realization of nanophotonic [6] and nanomechanic [7,8] devices in bulk diamond crystals will also be discussed. Finally, I will present our recent result on frequency comb generation at telecom wavelengths using high Q factor (>10^6) diamond ring resonator [9], and discuss our ongoing efforts towards realization of diamond comb in visible range. (http://nano-optics.seas.harvard.edu.)

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[1] T.M. Babinec et al., "A bright single photon source based on a diamond nanowire," *Nature Nanotechnology* **5**, 195 (2010).

[2] J.T. Choy et al., "Enhanced Single Photon Emission by Diamond-Plasmon Nanostructures," *Nature Photonics* **5**, 738 (2011).

[3] B.J.M. Hausmann et al., "Integrated Diamond Networks for Quantum Nanophotonics," *Nano Letters* **12**, 1578 (2012).

[4] B.J.M. Hausmann et al., "Coupling of NV centers to photonic crystal nanobeams in diamond," *Nano Letters* **13**, 5791–5796 (2013).

[5] M.J. Burek et al., "Free-standing mechanical and photonic nanostructures in single-crystal diamond," *Nano Lett.* **12**, 6084 (2012).

[6] M. Burek, Y. Chu, M. Liddy, P. Patel, J. Rochman, W. Hong, Q. Quan, M.D. Lukin, and M. Loncar, "High-Q optical nanocavities in bulk single-crystal diamond," *Nat. Comm.* **5**, 5718 (2014).

[7] M. Burek et al., "Nanomechanical resonant structures in single-crystal diamond," *Appl. Phys. Lett.* **103**, 131904 (2013).

[8] Y.I. Sohn et al., "Dynamic Actuation of Single-Crystal Diamond Nanobeams," *arXiv:1408.5822.*

[9] B. Hausmann et al., "Diamond Nonlinear Photonics," *Nature Photonics* **8**, 369 (2014).

WK1

Plasmonic Arrays

George C. Schatz

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Silver and gold nanoparticles have unique optical properties that are associated with the excitation of collective excitations of the conduction electrons known as plasmon resonances. The resonance frequencies are sensitive to particle shape and size, which means that the color of the nanoparticles can be tuned over a wide range of wavelengths, and they are also sensitive to the arrangement of the nanoparticles into aggregates and arrays. This talk will emphasize recent theory and experiments that have probed the effect of arrays of these particles in 1D, 2D and 3D on optical response. The arrays in 1D and 2D can be made using standard lithography tools, but much of the talk will emphasize bottom-up assembly of arrays that is possible using DNA-functionalized nanoparticles and self-assembly of nanoparticle superlattices driven by DNA hybridization. We show that the array structures lead to new kinds of hybrid optical modes in which localized surface plasmon resonances in the nanoparticles are coupled with photonic modes of the lattices, including Bragg modes, Fabry-Perot modes and other modes. These hybrid modes are often much narrower than the isolated particle plasmons, and films composed of these superlattices have unusual metamaterials properties. We also show that for 2D lattices it possible to generate a new class of sub-wavelength laser in which excitons in laser dyes are coupled with the hybrid lattice modes to produce enhanced stimulated emission.

WK1

Interfacial Influence on Surface Plasmon Resonances in "Quantum-sized" Metal Nanoparticles

Sheng Peng, Zheng Li, Jonathan J. Foley, IV, Stephen K. Gray, Gary P. Wiederrecht, and Yugang Sun

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Nanoparticles made of noble metals such as silver and gold exhibit strong optical absorption due to their surface plasmon resonance (SPR) that corresponds to the collective oscillation of surface conduction electrons in response to the incident electromagnetic waves. As the nanoparticles are smaller than 20 nm (i.e., the size in the range similar to that of quantum dots), the interfaces formed on the surfaces of these noble metal nanoparticles significantly influence the nanoparticles' SPR. For example, the monodispersed silver nanoparticles synthesized *via* a well-defined chemical reduction process exhibit an exceptional size-dependence of SPR peak positions: as particle size decreases from 20 nm the peaks blue-shifts but then turns over near ~12 nm and strongly red-shifts. Theoretic modeling and calculations reveal that the surface chemistry corresponding to the interactions between the capping molecules and the surface silver atoms in the nanoparticles become pronounced in determining their optical properties because the surface silver atoms represent a significant fraction of the total number of atoms in small nanoparticles. Such surface chemistry reduces the density of conduction band electrons (i.e., free electrons) in the surface layer of metal atoms, thus consequently influences the frequency-dependent dielectric constant of the metal atoms in the surface layer and the overall SPR absorption spectrum. In this presentation, a number of interfaces including the aforementioned metal/surfactant interface, metal/inorganic dielectric interface, and metal/metal interface will be discussed to highlight the importance of interface conditions on SPRs in "quantum-sized" noble metal nanoparticles. A comprehensive understanding of the relationship between interfacial coupling/chemistry and optical properties will be beneficial to exploit new applications of small colloidal metal nanoparticles, such as colorimetric sensing, electrochromic devices, surface enhanced spectroscopies, and photocatalysis.

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WK1

Crafting Nanophotonic Structures

Zhiqun Lin

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Colloidal nanocrystals exhibit a wide range of size and shape dependent properties and have found application in a myriad of fields such as photonics, optics, electronics, mechanics, drug delivery and catalysis to name but a few. Synthetic protocols that enable simple and convenient production of colloidal nanocrystals with controlled size, shape and composition are therefore of key general importance. Current strategies, however, often require stringent experimental conditions, are difficult to generalize, or require tedious multi-step reactions and purification. Recently, linear amphiphilic block copolymer micelles have been used as template for the synthesis of functional nanocrystals, but the thermodynamic instability of these micelles limits the scope of this approach. In this talk, I will elaborate general strategies for synthesizing a large variety of functional nanoparticles and nanorods (in particular, photonic nanoparticles and nanorods) with precisely controlled dimensions, compositions and architectures by using nonlinear block copolymers as templates. This new class of copolymers forms unimolecular micelles that are structurally stable under various experimental conditions and therefore overcomes the intrinsic instability of linear block copolymer micelles. Our approach enables the facile synthesis of organic solvent- and water-soluble nearly monodisperse photonic nanoparticles and nanorods with desired composition and architecture, including core/shell and hollow nanostructures.

WK1

SERS Inside Metal Nanoshell and Its Applications

Peng Zhang

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The observation of giant enhancement of Surface-enhanced Raman scattering (SERS) in the late 1990s had reignited the tremendous interests in the field. A great variety of nanostructures have been explored as SERS substrates, with various successes in terms of consistency and reproducibility. We approached the subject from a different aspect by placing the Raman probe molecules inside the metal nanoshell, in contrast to other previous experimental works, where SERS effect was studied only for molecules located in the proximal distance outside a nanostructure.

We show that enormous SERS enhancement inside metal (Au, Ag or mixed Ag/Au) nanoshell can be consistently and reproducibly achieved experimentally in the solution. The results provide new insights to the SERS effect, and pave ways for a new design of nanostructures to be used in Raman-based assays and imaging. Nanostructures of such design may also have profound implication in other plasmonic applications.

WK1

Multifunctional Diagnostic, Nanothermometer, and Photothermal Nano-devices

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In this study, the known therapeutic capabilities of gold nanorods (AuNRs) have been combined with the diagnostic and nanothermometer abilities of upconversion nanoparticles (UCNPs) to develop a system for simultaneous biological imaging, photothermal therapy, and nanothermal sensing. Both the excitation of UCNPs and the finely tuned longitudinal surface plasmon resonance (LSPR) mode of AuNRs lay in a window of relatively high light penetration of tissue in the infrared. The nanothermometer property of the UCNPs allows direct quantification of the localized temperature of the photothermally heated AuNRs chemically adsorbed to their surface and is free from the bleaching problems inherent in dye thermal sensing systems, especially at high laser fluences required to kill tissue. Spectroscopy on single particles, verified by transmission electron microscopy (TEM), has been performed at varying temperatures to confirm 1) the thermal sensing properties of UCNPs and 2) to finely tune their upconversion

enhancement arising from the LSPR coupling of the AuNRs. Preliminary quantification of the localized AuNR temperatures upon photothermal heating will be confirmed through single particle spectroscopy of the attached UCNPs. HeLa cell viability studies have also been performed.

WK1

Theoretical Studies of Hybrid Plasmon/Quantum-dot Systems

Stephen K. Gray

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I discuss classical electrodynamics and cavity quantum electrodynamics approaches to understanding and predicting the behavior of systems composed of metal nanoparticles and quantum dots such as CdSe nanocrystals. The plasmon resonance of the metal nanoparticle can significantly alter the radiative properties of the quantum dots and its correct incorporation into the modeling is emphasized. New avenues in nonlinear optical phenomena and quantum information are explored with these systems.

WK1

Photons in Flatland: Manipulating Light and Matter in Two-dimensional Nanomaterials

Nathaniel P. Stern

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Hybrid systems integrating light with matter offer a highly controllable landscape for understanding the interface between disparate physical entities. The emergence of materials with atomic-scale thickness suggests a new landscape in which to play with the coupling between light and low-dimensional materials. Exemplifying the interest of this new regime, the crystal symmetry of monolayer two-dimensional (2D) semiconductors can exhibit degenerate, yet distinct, valleys in momentum space that can be separately addressed by polarized light. In this talk, I will describe how these optical transitions in 2D semiconductors can be harnessed for manipulating electronic excitations and charge motion in two-dimensional materials. Drawing on the developments in cavity quantum electrodynamics of the last several decades, I will discuss the behavior of 2D semiconductors when integrated into optical cavities and photonic devices. I will use this approach to illustrate the potential for exploring new hybrid regimes of light-matter coupling based on engineering quantum interactions with nanoscale materials.